

# MULTI-STACKED InAs/GaAs QUANTUM DOT STRUCTURES AND THEIR PHOTOVOLTAIC CHARACTERISTICS

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## ABSTRACT

Multi-stacked InAs/GaAs quantum dot (QD) structures having various number of stacks were grown by molecular beam epitaxy (MBE) on  $n^+$ -GaAs substrate. They were fabricated into Schottky barrier devices for photovoltaic characterizations. QDs at a slow growth rate of 0.01 ML/s were chosen for all samples in the whole experiment. Current-voltage (I-V) curves under AM1 solar simulator were measured. It was found that larger number of QD stacks gives more short-circuit current. Contribution to photovoltaic effects by multi-stacked InAs QDs in these matrix materials was confirmed by spectral response measurement, which shows that apart from the sharp drop of photo-response at the band edge of GaAs ( $\sim 0.9 \mu\text{m}$ ), spectral peak at 1.1-1.2  $\mu\text{m}$ , corresponding to quantized energy of InAs QDs, was observed from these multi-stacked QDs samples. Photocurrent measurement was conducted at 1.1  $\mu\text{m}$  wavelength by a band-pass filtered light source to see the correlation between photocurrent at this specific long wavelength and the QD stack numbers.

## 1. INTRODUCTION

Self-assembled quantum dots (SA-QDs) are intensively investigated for several device applications [1-3] due to recent work on their improved dot quality and dot controllability [4]. Defect-free QDs could provide strong luminescence at room temperature [5] with reasonable sharp spectrum, which stems from their nature of quantized energy and high state density [6]. Therefore, SA-QDs are proposed to be utilized in an active layer of semiconductor lasers with very low threshold current density [7, 8]. In a reverse action, we are investigating that SA-QDs would be applicable for an active layer of photovoltaic cells with theoretically expected high efficiency [9].

We prepared InAs/GaAs SA-QDs by MBE technique and fabricated them into Schottky devices for electrical and optical characterizations. Schottky structure was selected in this work so that we could understand the intrinsic property of SA-QDs, and preparation of the samples with such a structure is simple as well. In photovoltaic applications, the advantages of using SA-QDs for photon absorption in the long wavelength part of solar spectrum have been discussed [10]. Multi-stacked Schottky QD solar cells with graded dot sizes were also realized [11].

QD size is controlled by varying the MBE growth rates. The QD structure was investigated by atomic force microscopy (AFM) and photoluminescence (PL) spectroscopy. The Schottky diodes incorporated with multi-stacked QDs were then realized by gold (Au)

evaporation. The photovoltaic effect is observed from these QD devices by current-voltage characterization at dark and under the AM1 solar simulator.

In this presentation, we fixed the QD size by using the same growth rate during the whole experiment but varying the number of stacks so as to observe the effectiveness of QD stacks for increasing photon absorbing layer. Short-circuit currents, especially at 1.1- $\mu\text{m}$  wavelength corresponding to quantized energies of InAs QDs, were reported and discussed for QD Schottky devices with different stack numbers.

## 2. EXPERIMENTAL PROCEDURE

The samples were grown by solid-source MBE on  $n^+$  GaAs (001) substrates. The doping concentration of the substrate was about  $5 \times 10^{18} \text{ cm}^{-3}$ . After desorbing the oxide at 630 °C under As pressure, a 1.0- $\mu\text{m}$ -thick GaAs buffer layer was deposited at 610 °C. The growth rate of GaAs was 0.6 monolayer per second (ML/s). During the growth, the beam equivalent pressure of  $\text{As}_4$  source was kept constantly at  $1.0 \times 10^{-5}$  mbar. The samples were then cooled down to 500 °C, which was previously calibrated by reflection high-energy electron diffraction transition from  $(2 \times 4)$  to  $c(4 \times 4)$  reconstruction of the GaAs surface. A 1.8-ML InAs SA-QD layer was deposited at this temperature. 0.01 ML/s InAs growth rate was used to produce SA-QDs. After finishing the growth of the QD layer, a 100-nm thick GaAs barrier layer was capped without growth interruption. This growth process was repeated to grow multi-stacked QD samples. The number of stacks was varied from 1, 2, 5 and 10. Overall sample structure is shown in Fig. 1.

After MBE growth, Au:Ge/Ni was deposited on the substrate by blanket evaporation, after which the samples were annealed in  $\text{N}_2$  flowing gas at 475 °C to form the ohmic contact. On the front side of the sample, 100-nm

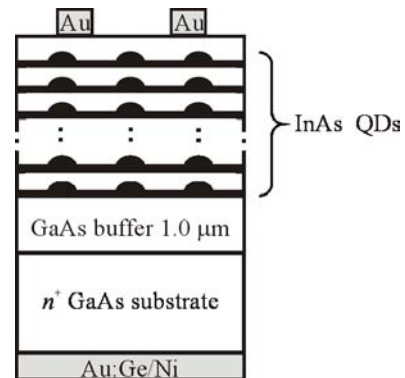


Fig.1 Sample structure of multi-stacked QDs.

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14. ABSTRACT <b>Self-assembled quantum dots by MBE (Molecular Beam Epitaxy) are examined as a structure of solar cells and other novel nanoelectronic devices. Zero-dimensionality of quantum dot gives rise to certain unique properties: charge storage and single-electron transport due to Coulomb blockade. The devices have low energy consumption and high speed operation due to quantized states inherent in quantum dot structure. Basic research on self-assembled quantum dots is detailed to understand the growth mechanisms and the electrical properties of the quantum dots so as to realize practical engineering applications like quantum dot solar cells, quantum dot lasers and single electron devices for quantum computing. The uniqueness of quantum dot solar cells is the possibility that they possess charge storage function, in addition to their electricity generating capability. Multi-stack with well controlled dot size is the key approach to be adopted throughout the research. Self-assembled quantum dots are very practical in engineering point of view. Another basic research would be the ordering of self-assembled quantum dots, which is an important requirement for many nanoelectronic devices.</b>					
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thick Au film was evaporated on to the surface through an E-shaped mask. The Au Schottky contact was used to realize the Schottky diodes with embedded InAs QDs. Finally, the samples were cleaved to reduce leakage currents along the edges. The surface area of each device is about  $2 \times 3 \text{ mm}^2$ .

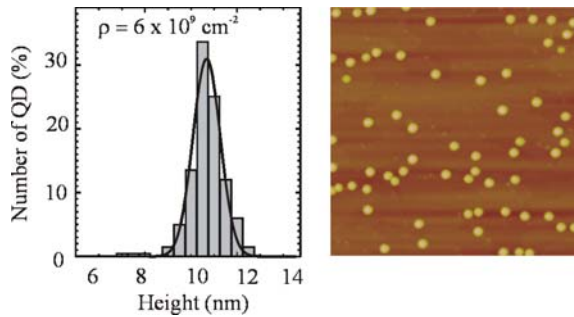
The current-voltage characteristics, observed in the dark and under the arc-lamp light source from a solar simulator, were carried out using the HP4140B pA meter. The constant AM1 light was calibrated from a standard reference Si solar cell. All I-V curves were confirmed by the results from a curve tracer.

To observe the contribution of SA-QDs in photovoltaic effects, photocurrent response at  $1.1 \text{ }\mu\text{m}$  wavelength were measured by filtered light source on SA-QD samples with different number of QD stacks.

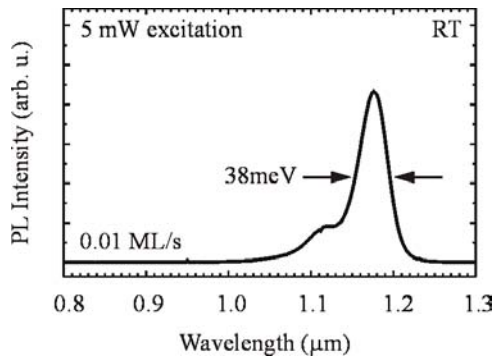
### 3. RESULTS AND DISCUSSIONS

Fig. 2 shows  $1 \times 1 \text{ }\mu\text{m}^2$  AFM image of 1.8 ML InAs/GaAs SA-QDs grown by  $0.01 \text{ ML/s}$  growth rate on the surface. Cross-sectional analysis by the AFM indicates that the typical height and diameter of QDs are  $10 \text{ nm}$  and  $60 \text{ nm}$ , respectively. The dot density is in the order of  $6 \times 10^9 \text{ cm}^{-2}$ . Fig. 3 shows the room-temperature PL spectrum having the peak at around  $1.17 \text{ }\mu\text{m}$ . PL linewidth ( $38 \text{ meV}$ ) agrees well with QD height distribution obtained from AFM analysis.

SA-QD Schottky samples with different number of QD stacks were characterized at dark and illuminated conditions for their I-V curves as shown in Fig. 4. I-V



**Fig. 2**  $1 \times 1 \text{ }\mu\text{m}^2$  AFM image of 1.8 ML InAs/GaAs SA-QDs grown at  $0.01 \text{ ML/s}$

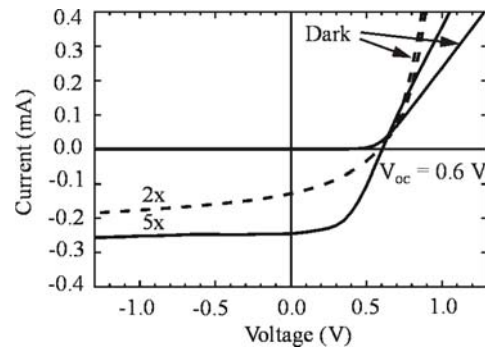


**Fig. 3** PL spectrum of InAs/GaAs SA-QDs embedded in GaAs at RT

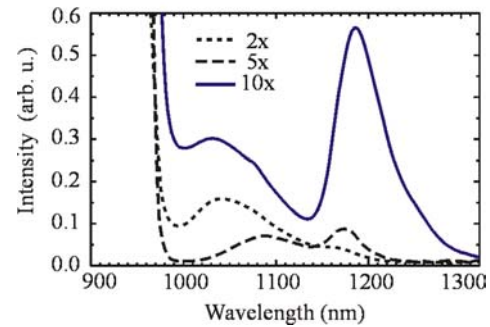
curves in the dark show typical rectifying contact of Schottky diodes. With AM1 illumination at  $100 \text{ mW/cm}^2$  intensity, the photovoltaic effect from the devices were observed in the fourth quadrant. Larger photocurrent could be expected and confirmed when the number of QD stacks is increased. The sample with 5 QD stacks shows typical results,  $V_{oc} = 0.6 \text{ V}$ ,  $J_{sc} = 9.54 \text{ mA/cm}^2$  and  $F.F. = 0.59$ , leading to a few percent efficiency for this non-optimized Schottky solar cell.

The spectrum response of these multi-stacked QD samples were measured by using chopped-beam tungsten-halogen lamp with an appropriate cut-off filter (IR 69) through a monochromator and a lock-in amplifier measuring system. The results are shown in Fig. 5, focusing at around  $1.1\text{-}\mu\text{m}$  wavelength spectra originating from the existence of the QDs.

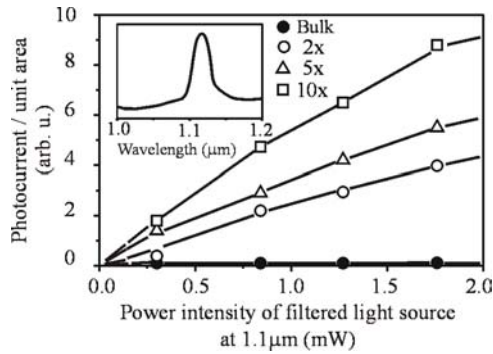
To confirm the long-wavelength response feature of QD samples at the specific wavelength of  $1.1 \text{ }\mu\text{m}$ , photocurrent measurements were carried out by using a tungsten-halogen lamp with a band-pass filter ( $1.1 \text{ }\mu\text{m}$ ) and a pA meter. The correlation between intensity dependence of photocurrents at  $1.1 \text{ }\mu\text{m}$  and the number of stacks is plotted in Fig. 6. The inset in Fig. 6 shows the spectrum of filtered light source having peak at  $1.12 \text{ }\mu\text{m}$ . This filtered source has narrow spectrum ( $\sim 40 \text{ nm}$ ) leading to low light power (only  $1\text{-}2 \text{ mW/cm}^2$ ) comparing to that of AM1 solar simulator. The spectral peak of filtered source is at  $1.12 \text{ }\mu\text{m}$  which has some mismatch to  $1.17 \text{ }\mu\text{m}$  of PL peak. These two experimental conditions lead to smaller photocurrents than expected from the QD samples. However, the experimental results clearly indicate that larger number of QD stacks gives higher photocurrent and bulk sample without QD stacks gives no photocurrent



**Fig. 4** I-V curves at dark and illuminated conditions of QD samples.



**Fig. 5** Spectral response at around  $1.1\text{-}\mu\text{m}$  wavelength of QD samples



**Fig. 6** Photocurrents from multi-stacked QD Schottky samples at 1.1- $\mu\text{m}$  wavelength comparing to bulk samples.

response at this specific long wavelength. It is noticeable that there is a trend of photocurrent saturation at larger number of QD stacks in these Schottky QD devices. The total thickness of 5 stacks InAs SA-QD structure including GaAs barriers (100 nm each) is about  $5 \times 100$  nm or 0.5  $\mu\text{m}$ . This saturation phenomenon is due to the nature of the Schottky structure whose junction is considered to be very shallow or zero-junction. Consequently, not all the QD stacks contribute to photovoltaic effect, and this limits the device performance if the Schottky structure is used.

The effectiveness of QD multi-stacked structure would be more pronounced in a p-n junction device where space-charge region could be controlled and designed to cover all the QD stacks so that all QDs could contribute to the photovoltaic effects. Further investigation on the QD structure under the internal field of p-n junction will be carried on in order to clarify this result. However, the Schottky structure in the present study gives us a basic understanding of QDs functioning in photovoltaic device.

#### 4. CONCLUSION

The multi-stacked QD Schottky devices with various QD stack numbers were fabricated by MBE and tested for the investigation of their photovoltaic characteristics. Large QDs at a slow growth rate of 0.01 ML/s was chosen for the whole experiment. The dot size and the dot density were identified by AFM image. The room-temperature photoluminescence results clearly reveal the quality and uniformity of the dots having quantized energy nature. Dark and illuminated I-V curves were measured on all QD samples. Multi-stacked QD structure gives more photocurrent due to thicker active layers of the device. Response of QDs to long wavelength particularly at 1.1  $\mu\text{m}$  was confirmed by spectrum response measurement. Photocurrents at the specific wavelength of 1.1  $\mu\text{m}$  were measured on all QD samples with different stack numbers. Larger number of QD stacks gives higher photocurrent but the photocurrent becomes saturated at large stack numbers due to the shallow junction of Schottky devices.

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#### REFERENCES

- [1] Y. Arakawa, "Progress in Self-Assembled Quantum Dots for Optoelectronic Device Application", *IEICE Trans. Electron.* Vol. **E85-C**, No. 1, 2002.
- [2] H. Ishikawa, "Applications of Quantum Dot to Optical Devices", *Semiconductors and Semimetals*, Vol. **60**, R.K. Willardson and A.C. Beer (eds.), Academic Press, London, 1999.
- [3] A.J. Shields, et al., "Single Photon Detection with a Quantum Dot Transistor", *Jpn. J. Appl. Phys.*, Vol. **40**, pp. 2058-2064, 2001.
- [4] Suwit Kiravittaya, "Homogeneity Improvement of InAs/GaAs Self-Assembled Quantum Dots Grown by Molecular Beam Epitaxy", Ph.D. Thesis, Chulalongkorn University, 2002.
- [5] Rudeesun Songmuang, Suwit Kiravittaya, Montri Sawadsaringkarn, Somsak Panyakeow, O.G. Schmidt, "Photoluminescence Investigation of Low Temperature Capped Self-Assembled InAs/GaAs Quantum Dots", *Journal of Crystal Growth*, 2003.
- [6] M. Asada, Y. Miyamoto, Y. Suematsu, "Gain and the Threshold of Three Dimensional Quantum-Box Lasers", *IEEE Journal of Quantum Electronics*, QE-**22**, pp. 1915-1921, 1986.
- [7] P.G. Eliseev, et al., "Ground-State Emission and Gain in Ultra low-threshold InAs-In GaAs Quantum Dot Lasers", *IEEE Journal on Selected Topics in Quantum Electronics* **7**, pp. 135-142, 2001.
- [8] I.Z. Alferov, Nobel Lecture, "The Double Heterostructure Concept and Its Applications", *Review of Modern Physics* **73**, pp. 767-782, 2001.
- [9] A. Marti, L. Cuadra, A. Luque, "Design Constraints of the Quantum-Dot Intermediate Band Solar Cell", *Physica E*, Vol. **14**, pp. 150-157, 2001.
- [10] V. Aroutiounian, et al., "Quantum Dot Solar Cells", *J. Appl. Phys.*, Vol. **89**, pp. 2268-2271, 2001.
- [11] S. Kamprachum, et al., "Multi-stacked Quantum dots with Graded Dot Sizes for Photovoltaic Applications", *Proceedings of 29<sup>th</sup> IEEE Photovoltaic Specialist Conference*, New Orleans, Louisiana, U.S.A., May, 2002.